

LA-UR--87-3283

DE88 000482

TITLE: DIELECTRIC CHANGES IN NEUTRON-IRRADIATED  
RF WINDOW MATERIALS

AUTHOR(S): H. M. Frost and F. W. Clinard, Jr.

SUBMITTED TO: Third International Conference on Fusion Reactor Materials,  
Karlsruhe, FRG October 4-8, 1987

Proceedings to be published in the Journal of Nuclear Materials

**DISCLAIMER**

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy

---

**MASTER**  
**Los Alamos** Los Alamos National Laboratory  
Los Alamos, New Mexico 87545

# Dielectric Changes in Neutron-Irradiated RF Window Materials

H. M. Frost and F. W. Clinard, Jr.

Materials Science and Technology Division

Los Alamos National Laboratory

Los Alamos, NM 87545, USA

## Abstract

Ceramics used for windows in ECRH heating systems for magnetically-confined fusion reactors must retain adequate properties during and after intense neutron irradiation. Of particular concern is a decrease in transmissivity, a parameter inversely related to the product of dielectric constant  $K$  and loss tangent  $\tan\delta$ . Samples of polycrystalline  $\text{Al}_2\text{O}_3$  and  $\text{BeO}$  were irradiated to  $1 \times 10^{26} \text{ n/m}^2$  at 660K in the EBR-II fission reactor, and the above properties subsequently measured at 95 GHz. It was found that  $K\tan\delta$  for both materials doubled, implying a doubling of thermal stresses and a consequent reduction of time-to-failure from an assumed one year to 20 min for beryllia and 2 s for alumina. In the case of  $\text{BeO}$ , a large increase in reflectance of the incident millimeter-wave power results from dielectrically uncompensated swelling. This phenomenon could significantly degrade source performance.

## 1. Introduction

Electron cyclotron resonant heating (ECRH) is one of the techniques available to raise the temperature of a magnetically-confined plasma to ignition. Development of a window to isolate the RF source and its

SF<sub>6</sub>-pressurized waveguide from the plasma presents a severe problem for designers and for ceramic science.

In an ideal system the window would be perfectly transparent to the millimeter waves from the source, and would then have only to survive the abuses that can result from the irradiation field (e.g., swelling, strength loss, reduction of thermal conductivity). In fact, however, the window will suffer heating from absorption of a portion of the incident power, and this can lead to thermal stresses sufficient to fracture the window even without degradation of mechanical and thermal properties.

Window designs for ECRH applications are likely to include a coolant flowing between two parallel ceramic slabs. Under these conditions and assuming realistic operating parameters, Fowler [1] has shown that large stresses can indeed be generated. Heat deposited in the window is in fact approximately proportional to loss factor (the product of dielectric constant K and loss tangent tanδ), as shown in the relationship:

$$Q \propto \omega \epsilon_0 K \tan \delta E_0^2 \quad (1)$$

where  $\omega = 2\pi f$ ,  $\epsilon_0$  is the permittivity of free-space vacuum, and  $E_0$  is the rms electric field amplitude. Materials properties of concern include not only loss factor but strength, thermal conductivity, and density (swelling). The work described here concentrates on the 'least-known' K and tanδ, reporting changes induced in Al<sub>2</sub>O<sub>3</sub> and BeO by neutron irradiation to a level likely to be attained in service. The results are used to predict the effect of irradiation on window lifetime and on reflective ("return") losses, and are discussed in terms of overall materials and system performance.

## 2. Experimental procedure

Specimens of alumina and beryllia were irradiated to a fluence of  $1 \times 10^{26}$   $n/m^2$  ( $E > 0.1$  MeV) at 660K in the EBR-II fast fission reactor. Control specimens were annealed at the same temperature and for the same number of full-power days. The alumina was Coors AD-995 (nominally 99.5% pure), the beryllia a BeO-based ceramic containing significant amounts of Al and Si. Major impurities (in wt.ppm unless otherwise indicated) were: for the alumina, 1000 Si, 700 Ca, 600 Mg, and 200 Fe; for the beryllia, 1-10% Al, 1% Si, 2000 Mg, 600 Fe, and 300 Ca.

Swelling values were obtained by direct measurement of specimen masses and dimensions. Dielectric properties were determined by use of a millimeter wave (MMW) measuring apparatus [2], a block diagram of which is shown in Fig. 1. In this system an IMPATT diode generates the MMW, which is swept from 90 to 100 GHz by the sweeper. These signals impinge on a precisely-dimensioned waveguide-enclosed sample, where they are partially transmitted, absorbed, and reflected. Fig. 2 shows the specimen and waveguide arrangement. Nominal inside dimensions of the WR-10 waveguide used are 1.26 mm by 2.54 mm. The specimens are dimensioned for a 'push-and-slide' fit inside the waveguide.

The procedure used to obtain data from irradiated and control specimens is described elsewhere for low-loss materials [2]. Briefly, amplitudes of the signals were extracted by high-directivity couplers isolated (by isolators) from their corresponding square-law detectors and displayed logarithmically as either reflection loss (RL) or transmission loss (TL). From the spacings between successive RL-null frequencies corresponding to successive resonances in the specimen length, one can deduce  $K$ ;  $\tan \delta$  is directly proportional to the

TL values at these same frequencies. Both the RL and TL spectra entail differences between measurement and calibration curves, the latter involving reflections from a high-reflectivity metal plug and transmission through the empty waveguide for RL and TL, respectively. Various special techniques were adopted to reduce experimental error and improve reproducibility of results [2].

### 3. Results and discussion

#### 3.1. Window lifetime

Changes in density, dielectric constant, and loss tangent for the two test ceramics are shown in Table 1. Both materials swelled significantly, with results for alumina being in agreement with those obtained by Tucker et al. [3] for other specimens of this material exposed to the same irradiation conditions. Values for beryllia are consistent with those reported by Hickman [4]. These dimensional changes are discussed here only in the context of their effect on dielectric properties of the window; however, it should be recognized that unless a compliant window-mounting system can be devised swelling can induce severe mismatch stresses.

Starting dielectric properties for both control materials are consistent with published values. Relative to the control samples, irradiation caused a slight decrease in dielectric constant but more than a doubling of loss tangent. From Eq. (1) a doubling of  $K \tan \delta$  in either material will lead to a doubling of heating rate, and that in turn will lead to a doubling of window hoop stress [1]. (We assume in this analysis that loss factor changes yield

proportional changes in absorption.) Because of the extreme sensitivity of time-to-failure to stress for fracture statistics based on the Weibull distribution, a marked reduction in lifetime of RF windows can be expected. As an example, we have used formulations and data of Ferber et al. [5] to estimate lifetimes for alumina and beryllia windows of 3.175 cm radius (exposed face) subjected to 102 kW and 56 kW continuous power, respectively, in the  $TE_{02}$  mode (circular waveguide) at 100 GHz. Assuming conditions where loss factor is doubled and inert tensile fracture strengths are 362 MPa for  $Al_2O_3$  [6] and 137 MPa for BeO [5], time-to-failure for a window failure probability of 0.01 (1 window in 100) drops from 1 yr to 20 min for BeO, and from 1 yr to just 2 s for  $Al_2O_3$ .

These results take on added meaning if viewed in two alternative ways. First, as might be guessed from Eq. (1), window lifetime can be restored to one year if the power loadings are halved. Second, to reach the 200 kW loading that has been identified as a desirable goal [5], loss tangent for both alumina and beryllia could be no greater than  $\approx 10^{-4}$  during use.

The above observations assume a constant value of thermal conductivity, whereas in reality that property is likely to be markedly degraded in both test materials by neutron damage [4,7]. Since under steady-state heating conditions the heat source term in the heat conduction equation is inversely proportional to this parameter [5], a halving of thermal conductivity has the same effect as a doubling of loss tangent. In other words, a quadrupling of thermal stresses can reasonably be expected under the test conditions used here if degradation of thermal conductivity is taken into account, and window performance would be degraded still further.

Strength of non-cubic ceramics such as  $Al_2O_3$  and BeO is readily

reduced by neutron irradiation, as anisotropic swelling causes high internal strains and microcracking. For alumina irradiated under the conditions employed here, strength is degraded by 25% [3], whereas degradation is likely to be much worse for BeO [4]. And so again, window performance can be expected to suffer more than that shown in the above calculations.

Considering the deleterious effects of increase in loss factor, reduction of thermal conductivity, and decrease in strength for these two test materials, it is apparent that placement of the RF window in a lower radiation flux than that employed here or selection of a more radiation-resistant ceramic must be considered. (The dose used here is equivalent to about 6 months' exposure at the first wall of a  $2 \text{ MW/m}^2$  fusion device.) Future designs may specify a more remote location, but must take into account the degraded power-handling performance of the section of waveguide between window and first wall if that section can no longer contain  $\text{SF}_6$  dielectric gas.

From the standpoint of mechanical reliability, an optimal ceramic should have high strength, high thermal conductivity, and low loss factor as well as good retention of these properties under irradiation. Spinel ( $\text{MgAl}_2\text{O}_4$ ) shows good irradiation stability, but its starting properties are generally inferior to those of  $\text{Al}_2\text{O}_3$  [7]. A better possibility might be AlON ( $\text{Al}_{23}\text{O}_{27}\text{N}_5$ ), a ceramic with the spinel crystal structure and good starting dielectric and other properties [8]. Another approach is to improve the properties of present candidate materials; possible approaches include control of grain size and grain boundary phases, and addition of fine dispersed phases to reduce swelling or improve retention of initial thermal conductivity [7].

The work presented above focused on the role of increased loss factor as observed after irradiation. It should be emphasized that measurements made

during irradiation may show even greater degradation of this parameter, and that such tests are needed before definitive results can be obtained for use in materials selection and window design.

### 3.2. Reflection of the incident beam

Detuning of the window thickness  $d$  from the resonant condition of an integral number of half-wavelengths  $\lambda/2$  via radiation-induced swelling or changes in dielectric constant  $K$  may adversely affect the operation of some sources by increasing the millimeter-wave power reflected from the window back to the source. The concomitant changes in the impedance conditions could degrade the performance of, or perhaps even damage, the source, depending on type. A 1% linear swelling of a  $3(\lambda/2)$  thickness  $d$  would detune an alumina window's power-flow reflection coefficient  $R$  from the desired zero value to about 0.02, assuming  $K$  does not change. For comparison, this is about 200 times larger than the effect of thermal expansion alone due to a 100K temperature rise from room temperature, and four times larger than for a 1% change in  $K$  only. The detuning effect would be exacerbated if there is an accompanying radiation-induced increase in dielectric constant, but can be lessened if  $K$  decreases as observed here (Table 1).

Table 2 presents calculated values of reflection-related quantities for MMW materials of window thickness  $3 \lambda/2$  and a frequency of 95 GHz, in the control and irradiated condition. For purposes of reference, the  $R$  value of 0.02 referred to in the preceding paragraph corresponds to a reflection loss  $RL$  of -18 dB and a voltage standing wave ratio  $VSWR$  of 1.3. Calculations for control materials take their small values of  $\tan \delta$  into account to estimate the



tiny amount of power actually reflected from a low-loss window of nominally resonant thickness, and thus provide a baseline for comparison with irradiated window performance.

The trends go as follows: the larger the value of  $R$  (i.e., the closer it is to unity), then the closer to zero the value of  $RL$  or the greater the departure of VSWR from unity, and accordingly the greater the power reflected from the window back toward the source. Values of thickness swelling plus those for  $K$  and  $\tan\delta$  were substituted into standard formulas [9], or results derived from them, for calculation of the tabulated quantities.

These results may be interpreted as follows. If an irradiated alumina disk were used as a MMW window, over an order of magnitude more power would be reflected toward the source compared with the non-irradiated case, but this would still result in the ideal VSWR value of 1.0. The case is different for beryllia, however. The reflected power is roughly four orders of magnitude greater than that for the control, causing the VSWR to change from 1.0 to 1.8 for a single window (and to an estimated 2.3 for a double window) -- a matter of concern for performance of gyrotrons and other source components as well as increased likelihood of dielectric breakdown in the feed waveguides (because of increased electric fields resulting from the standing waves) and diminution in the power available for plasma heating. The oppositely-directed effects of swelling and reduction of dielectric constant of roughly the same magnitude do not result in a small net effect because of the approximate dependence of  $R$  on  $[(\delta d/d) + (1/2)(\delta K/K)]^2$ . However, in contrast to the case of window heating, reduction of radiation damage to  $K$  and  $d$  may be more amenable to the simple solution of selecting other commercially-available material grades. This is because the irradiation-induced changes in  $K$  and  $d$  are one to two orders of

magnitude smaller in effect than are the changes in  $\tan\delta$ . At any rate, when stability of the window's resonant thickness condition is added to the criteria already mentioned in Sec. 3.1 for mechanical reliability, an optimal RF window's linear swelling needs to be equal to 1/2 of its fractional change in dielectric constant,  $-(1/2) (\delta K/K) = \delta d/d$ .

### Conclusions

1. The present work represents the first systematic study of changes in dielectric properties of ceramics following high-dose fast-neutron damage.
2. Dielectric losses in both  $\text{Al}_2\text{O}_3$  and BeO were doubled by neutron irradiation, with most of the change attributed to increases in loss tangent.
3. Calculations show that lifetime of  $\text{Al}_2\text{O}_3$  and BeO windows can be decreased by seven and four orders of magnitude, respectively, as a result of beam heating and resulting thermal stresses.
4. Materials performance will be even further degraded by irradiation-induced reductions in strength and thermal conductivity.
5. Swelling-induced reflection of MMW power back to the source may in the case of irradiated BeO be great enough to present severe operating problems for ECRH systems.
6. Materials lifetimes will depend strongly on starting properties of the ceramics chosen for this application.
7. Future work should address:
  - the distinction between processes of absorption (leading to material heating) and scattering (which does not result in material heating but does contribute to  $\tan\delta$ ).

- the need for dielectric property data taken concurrently with irradiation.
- the necessity for an understanding of the nature of the defects (e.g., point, line, planar) responsible for changes in dielectric properties.
- the need for data on the effect of neutron damage on structural property considerations such as intragranular vs. grain-boundary damage, changes in Weibull modulus, and static fatigue behavior.
- the requirement that dielectric property data be sufficiently accurate and precise to allow at least order-of-magnitude 'accuracies' in predictions of window lifetimes.

#### References

1. J. D. Fowler, Jr., J. Nucl. Mater. 122 & 123, (1984) 1359.
2. H. M. Frost, in: Eighth Annual Progress Report on Special Purpose Materials for Magnetically Confined Fusion Reactors, U.S. Department of Energy Report No. DOE/ER-0113/5 (1986) p.11.
3. D. S. Tucker, T. Zocco, C. D. Kise and J. C. Kennedy, *ibid.*, p. 9.
4. B. S. Hickman, "Radiation Effects in Beryllium and Beryllium Oxide", in: Studies in Radiation Effects Series A; Physical and Chemical Vol.1 (ed. G. J. Dienes) (Gordon and Breach, New York, 1966) p. 72.

5. M. K. Ferber, H. D. Kimrey and P. F. Becher, J. Mater. Sci. 19 (1984) 3767 and P. F. Becher and M. K. Ferber, J. Mater. Sci. 19 (1984) 3778.
6. Ferber and Becher, 1987 (private communication).
7. F. W. Clinard, Jr., J. Mater. for Energy Systems 6 (1984) 100.
8. H. M. Frost and C. D. Kise, in: Fusion Reactor Materials Semiannual Progress Report for Period Ending September 30, 1986, U.S. Department of Energy Report No. DOE/ER-0313/1 (in press 1987).
9. M. Born and E. Wolf, Principles of Optics (McMillan, New York, 1964) pp. 61-66 and 627-632.

Table 1. Millimeter wave dielectric properties for neutron-irradiated ceramics, corrected for nonlinear  $f$ -vs.- $\delta f^*$  relationship for waveguide case

Material and Condition	Volume Swelling (%)	K	$\tan\delta$ ( $10^{-4}$ )	$K\tan\delta$ ( $10^{-3}$ )
---------------------------	------------------------	---	-------------------------------	--------------------------------

$Al_2O_3$

Control	--	9.86	3.9	3.8
Irradiated	+3.0	9.68	8.3	8.1

BeO

Control	--	6.23	6.5	4.0
Irradiated	+9.4	5.42	15.0	8.3

\* $f$  = frequency.

Table 2. Window detuning effects at 95 GHz

Material	R (Units of $10^{-3}$ )		-RL (dB)		VSWR	
	Control	Irradiated	Control	Irradiated	Control	Irradiated
$\text{Al}_2\text{O}_3$	0.0067	0.13	52	39	1.0	1.0
BeO	0.010	82	50	11	1.0	1.8

Note: Control:  $\tan\delta > 0$  assumed in calculations; irradiated:  $\tan\delta = 0$  in calculations. R = power-flow reflection coefficient, RL = reflection loss, and VSWR = voltage standing wave ratio. Baseline specimen thickness:  $3\lambda/2$ . Waveguide values of K and  $\tan\delta$  used for free-space calculations.

### Figure Captions

Fig. 1. The millimeter-wave measurement system. The block denoted CERAMIC is depicted in Fig. 2.

Fig. 2. Partial view of a waveguide section with specimen partially inserted, plus an exposed view of a separate specimen.

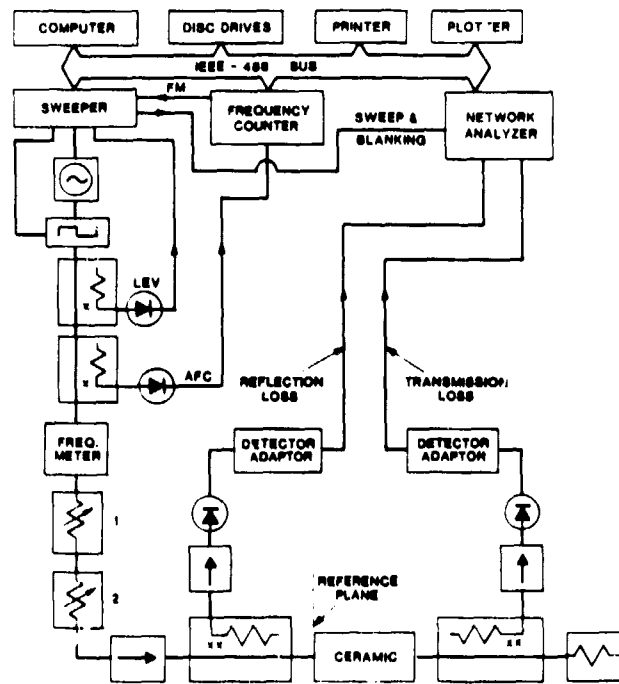


Figure 1

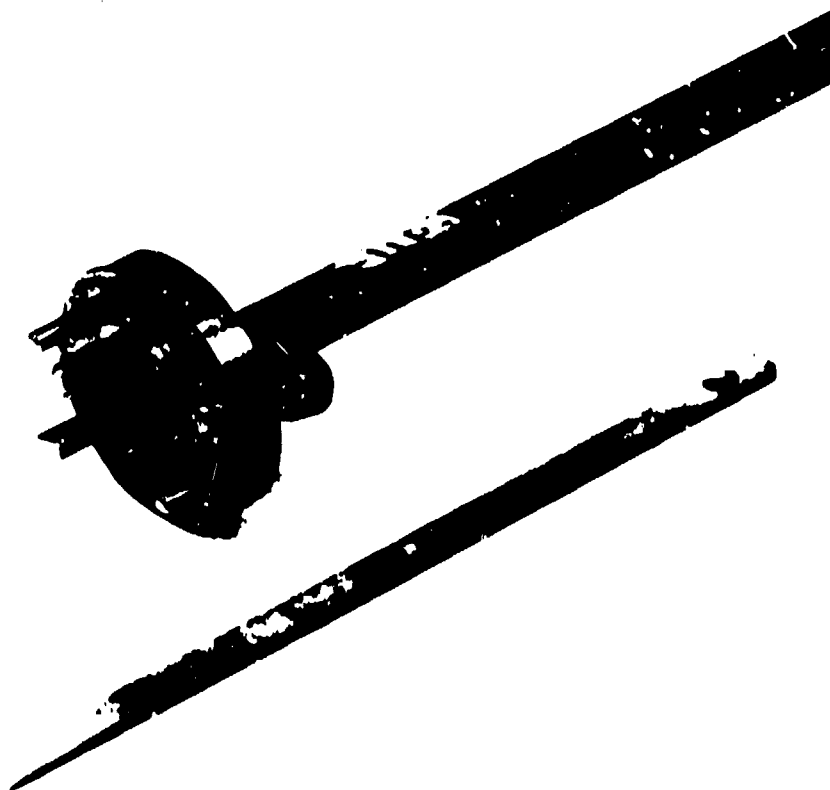


Figure 2